Response to Reviewers

Reviewer #2

Wang et al. present an analysis of VOC emissions measured from vehicle dynamometer testing for vehicles designed under different emission standards (China I - IV). The authors evaluate total and speciated VOC emissions from both gasoline, diesel, and LPG under a variety of conditions (cold start, warm start, speed, etc). The authors detail the different emission factors between each vehicle, and observe a distinct difference between the OVOCs emitted by gasoline and diesel engines. The latter produces significantly higher fraction of OVOCs than by gasoline, which appears to be at least partly associated with the pollution control technology.

I found the paper to be very well-written, well-reasoned, and full of good information. I appreciated the study as a nice piece of work describing fossil fuel emissions from motor vehicles in China.

My only substantive comment is that I don't have a sense of the fuel composition and how this might contribute to the high OVOCs observed in diesel exhaust. And how do the OVOC emissions compare against diesel exhaust studies reported elsewhere? Gentner et al. (2013) also see elevated OVOC emissions in diesel compared to gasoline. Are these differences comparable to what is observed here, or is there something different between the aftertreatment or fuels that could contribute to any differences?

Reply: We would like to thank the reviewer for the insightful comments, which helped us tremendously in improving the quality of our work. Fuel composition is one of determining factor for VOCs emissions from vehicles (Gentner et al., 2017). We conducted some literature review and added a discussion in Section 1 in the Supplement to provide some information about chemical compositions of gasoline and diesel fuel in China. We also appreciate the reviewer for providing the useful reference. We added some discussions in the Section 3.1 to compare with the result of them.

We also used the fractions of OVOCs in total VOC emissions to compare against diesel exhaust studies reported elsewhere (Fig.12). If only considering carbonyls among various types of OVOCs measured by PTR-ToF-MS, the OVOC fractions determined in this study are more comparable with previous studies. We also discussed higher OVOC emissions in diesel vehicles and impact on after-treatment devices, please find the response to individual comments below.

The sentence in the Section 2.1 (line 126-127) is modified to:

The detailed information for test vehicles is summarized in Sect. 1 in the Supplement, Table S2 and Table S3.

The Section 1 in the Supplement is modified to:

Fuel composition is one of determining factor for VOCs emissions from vehicles (Gentner et al., 2017). The gasoline fuel used in China is mainly comprised of C_4-C_7 hydrocarbons. The chemical compositions of gasoline fuel are alkanes (55%-62%), alkenes (12%-17%), aromatics (27%-32%), and methyl tert-butyl ether (MTBE, 1%-4%) (Tang et al., 2015;Sun et al., 2021;Qi et al., 2021;Huang et al., 2022). Heavy hydrocarbons, namely C_8-C_{10} alkanes and aromatics, contributed most in diesel fuel. The chemical compositions of diesel are alkanes (70%-79%), alkenes (1%-7%), and aromatics (21%-25%) (Wang et al., 2015;Yue et al., 2015;Hou and Jiang, 2018;Liu and Zhang, 2015). Gasoline and diesel fuel are summer blends, and the gasoline fuel does not content ethanol in this study.

The sentences in the Section 3.1 (line 284-289) are modified to:

As the largest OVOCs emitted from gasoline vehicles (4.6 ± 5.1 mg·km^{-1}), methanol is found to be the only common OVOC species, with lower emission factors from diesel vehicles than gasoline vehicles. The emission factor of other OVOCs (e.g., formaldehyde, acetone) from diesel vehicles are higher than gasoline vehicles, which is consistent with previous results (Gentner et al., 2013).

Comments:

1. Line 70-71 Based on the reference, I presume that the authors are specifically noting the decline of VOCs in urban regions in China? For clarity, I would suggest re-writing this sentence to say "Furthermore, VOC emission significantly decreased in China due to stricter emission standards."

Reply: We thank the reviewer for the comment. We corrected this sentence as **"Furthermore, VOC emissions from vehicles significantly decreased in China due to stricter emission standards (Liu et al., 2017;Sha et al., 2021)".**

2. Line 76: Could the authors provide some context on the China VI emission standard? I recognize that the standard is dependent on power ranges, but a few sentences on VOC emissions at max power output would be useful. This would also be useful in the methods (lines 112 - 122) to give readers context as to what the China I - IV standards represent in terms of VOC emissions.

Reply: We thank the reviewer for the comment. China VI emission standard is the newest emission standard for vehicles. Limits on exhaust emissions of gasoline vehicles are tightened by 30% to 50% from China V to China VI for different pollutants (Lyu et al., 2020). China VI emission standard is mainly reflected in the requirements for the emission limits of pollutants (e.g. CO, NO_x, THC, etc.). In response to the reviewer's question, power ranges and max power outputs are not directly reflected in China VI emission standard, and rarely reported in previous studies.

We added a discussion in Section 1 in the Supplement to provide some information about the China VI emission standard of vehicles. We added Table S7 with the different emission standards (China I - China VI) for light-duty vehicles (LDV) in gasoline and diesel as fuel, and Table S8 for heavy-duty diesel engines (HDDE) in different emission standards (China I - China V). We also added some discussion in the Section 2.1 to provide the averaged fractions of gasoline and diesel vehicles with different emission standards for the vehicle fleet in China, which is shown in the Table S1 in the revised manuscript (Table S6 in the original manuscript).

The sentences in the Section 1(line 76-78) are modified to:

The emission limits for various air pollutants emitted by vehicles are significantly lower under the China VI emission standard (see details in the Supplement) (Wu et al., 2017).

The Section 1 in the Supplement is modified to:

The limits and measurement methods for emissions of light-duty vehicles (GB18352.6-2016; known as the China VI standard) are introduced in the recent years in China, which applies to light-duty vehicles by gasoline or diesel as the fuel. The China VI emission standard continued the EU standard system as the reference with various regulation details integrated from US emissions standards (Lyu et al., 2020). Vehicle emission limits are significantly lower for the China VI standard (Table S7, Table S8). For example, limits on gasoline vehicle exhaust emissions were tightened by 30 to 50% from China V to China VI, and a new particulate number (PN) limit was added in gasoline vehicles (Lyu et al., 2020).

The sentence in the Section 2.1 (line 123-124) is modified to:

The averaged fractions of gasoline and diesel vehicles with different emission standards for the vehicle fleet in China are shown in Table S1 (MEEPRC, 2019;Li et al., 2021).

3. Line 81: Would suggest modifying "group" to say "class of compounds"

Reply: We replaced "group" with "class of compounds".

4. Line 80-83: Are the authors primarily discussing VOC measurements from dynamometer studies, or tunnel studies, or ambient studies? I think the distinction matters given that results from laboratory, tunnel, or ambient measurements can be interpreted differently given differences in co-emitted sources that can convolute the measured signal from tailpipe emissions

Reply: We thank the reviewer for the comment. We modified this sentence in the Section 1 to make it clearly in different vehicle measurement methods.

The sentence in the Section 1(line 82-86) is modified to:

Oxygenated volatile organic compounds (OVOCs) were found to be an important class of compounds in vehicle exhausts, accounting for more than 50% of the total VOC emissions for diesel vehicles from both chassis dynamometer tests (Schauer et al., 1999;Mo et al., 2016) and on-road mobile measurements (Yao et

al., 2015).

5. Lines 268-271: Are there also differences in the aftertreatment that might lead to higher OVOC emissions? The authors note the temperature of the device at line 240, and I'm curious if previous work has looked at VOC speciation under different aftertreatment conditions.

Reply: We thank the reviewer for the comment. After-treatment devices in vehicles have been improved associated with the upgrading of emission standards. According to the #8 comment of the reviewer, our results (Fig. 7c-d in the revised manuscript) actually can answer the question of the reviewer in this comment. The two graphs show that the chemical compositions of VOC emissions are comparable between different emission standards for both gasoline and diesel vehicles (R=0.98 and 0.89), indicating after-treatment devices may not affect the relative fraction of VOC components. We added some description and reference in the section 3.2, and added a discussion in Section 1 in the Supplement to provide some information about the after-treatment devices in gasoline and diesel vehicles.

The sentence in the Section 3.2 (line 363-366) is modified to:

Fig. 7c-d show that the chemical compositions of VOC emissions are comparable between different emission standards for both gasoline and diesel vehicles (R=0.98 and 0.89), indicating after-treatment devices may not affect the relative fractions of VOC components.

The sentence in the Section 3.2 (line 379-383) is modified to:

These results indicate the after-treatment device for diesel vehicles (see Sect. 1 in the Supplement for details.) may effectively reduce emissions of some heavier VOC species, though the after-treatment devices do not aim for VOCs control (Gentner et al., 2017).

The Section 1 in the Supplement is modified to:

After-treatment devices commonly used in light-duty gasoline vehicles are three-way catalyst (TWC) and gasoline particulate filter (GPF). They have been

improved with the upgrading of emission standard. For diesel vehicles, typical after-treatment devices include diesel oxidation catalyst (DOC), diesel particulate filter (DPF), and selective catalyst reduction (SCR) (Zhou et al., 2019;Lyu et al., 2020;Shen et al., 2021). The diesel vehicles for China III or prior do not have any after-treatment devices. Light-duty-diesel-truck (LDDT) used DOC and DOC+DPF as after-treatment devices in China IV and V diesel vehicles, respectively. SCR devices are mainly used for heavy-duty-diesel-truck (HDDT) with China IV and V as after-treatment devices.

6. Figure 1: It would be useful to see the acronyms (LDDT, MDDT, HDDT, and BUS) defined in the caption as a reminder to the reader.

Reply: We thank the reviewer for the comment. We added some description in the caption of Fig. 1about the acronyms (LDDT, MDDT, HDDT, and BUS). We also checked throughout the manuscript, and corrected the caption of Fig.2.

The caption of Fig.1 is modified to:

Figure 1. Real-time concentrations of acetaldehyde, acetone, benzene, toluene, and CO_2 for (a) a gasoline vehicle with emission standard of China I and (b) a light-duty diesel vehicle (LDDV) with emission standard of China IV. The two vehicles were both cold started. The gray shadows represent the speed of the vehicles on the chassis dynamometer.

The caption of Fig.2 is modified to:

Figure 2. The determined average mileage-based emission factors ($mg{\cdot}km^{-1}$) for (a) benzene, (b) toluene, (c) acetaldehyde, and (d) acetone for vehicles with different emission standards. The numbers above the top axis represent the number of all experiments (including multiple measurements for individual test vehicle) for each emission standard. LDDT, MDDT, HDDT, and BUS represent light-duty-diesel-truck, middle-duty-diesel-truck, heavy-duty-diesel-truck, and bus, respectively. Error bars represent standard deviations of emission factors for the specific emission standard.

7. Title of Section 3.2: The title doesn't quite reflect the discussion that follows. Might I suggest "Analysis of PTR-ToF-MS mass spectra to evaluate VOC speciation"?

Reply: We corrected this title in "Analysis of PTR-ToF-MS mass spectra to evaluate VOCs speciation".

8. Lines 320-323: This is a nice result, and partially addresses my question at lines 268-271. Could the authors point to this figure and discussion to demonstrate that the changes to the VOC distribution isn't significantly different between cold start and normal operation?

Reply: We thank the reviewer for the comment. After-treatment devices have been improved with the upgrading of emission standard. Our results (Fig. 7c-d in the revised manuscript) show that the chemical compositions of VOC emissions are comparable between different emission standards in gasoline and diesel vehicles ($R=0.98$ and 0.89), indicating after-treatment devices may not affect the relative fraction of VOC components.

Cold start is a major emission source of gasoline vehicles, which occurs after several hours of non-operation of vehicles (Gentner et al., 2017;George et al., 2015). Our results (Fig. 7a-b in the revised manuscript) demonstrate that variation behaviors are similar for different species and thus chemical compositions of VOC emissions are comparable between different start conditions. As cold start emissions are richer in unburned fuel than other hot-running conditions, the observation in Fig. 7a-b also infer that unburned fuel are the major contributor for vehicle exhaust emissions, which has been previously shown in California, US (Gentner et al., 2013). We added some discussions in 3.2 and Section 1 in the Supplement to provide some information about cold start in gasoline and diesel vehicles.

The sentence in the Section 3.2 (line 247-249) is modified to:

It might be a combined effect of cold engine and operation temperature of the after-treatment device (Gentner et al., 2017;George et al., 2015).

The sentences in the Section 3.2 (line 345-353) are modified to:

We observe strong correlation between emission factors from cold start and hot start tests (R=0.99 and 0.92) and generally consistent ratios between cold start and hot start for different types of VOC species for both gasoline and diesel vehicles, indicating that variation behaviors are similar for different species and thus chemical compositions of VOC emissions are comparable between different start conditions. As cold start emissions are richer in unburned fuel than other hot-running conditions, the observation in Fig. 7a-b also infer that unburned fuel are the major contributor for vehicle exhaust emissions, which has been previously shown in California, US (Gentner et al., 2013).

The Section 1 in the Supplement is modified to:

Cold start, which occurs after several hours of nonoperation for vehicles (Drozd et al., 2016), is a major source of emissions for gasoline vehicles and have greater emissions due to two issues: (1) low engine temperatures lead to incomplete combustion that allow non/partially combusted fuel compounds to exit engine cylinders. (2) Effective operation of the catalytic converter requires a warm-up period to reach sufficient catalyst operating temperatures (Gentner et al., 2017;George et al., 2015). Due to diesel emissions have emphasized control of primary $PM_{2.5}$ and NO_X emissions, the after-treatment devices of diesel vehicles (e.g. DOC, DPF, SCR etc.) do not aim for VOCs control.

9. Lines 424-425: I like the discussion in this section on using the aromatics to delineate between diesel and gasoline. I agree with the authors that these ratios might be difficult to assess in the ambient owing to additional sources of aromatics (e.g. solvent emissions) and secondary production of formaldehyde and acetaldehyde. Are there any unique masses, with high enough signal in ambient air, that could be used to more definitively separate gasoline vs diesel emissions? I also wonder if ratios to CO or other combustion markers might be insightful.

Reply: We thank the reviewer for the comment. Per the reviewer's comment, we have not found any other unique masses with high enough signals in gasoline or diesel vehicles that can be used for distinguish the two types of vehicles.

Furthermore, we added a Figure (Fig. S11b) with the emission ratios to CO (ppb·ppm^{-1}) between gasoline and diesel vehicles. The result (slope=0.16) is similar to the plot of emission factors between gasoline and diesel vehicles. A limited number of VOC species, including C_6-C_{10} aromatics are associated with higher emission ratios from gasoline vehicles, whereas the obtained emission ratios of most VOC species emitted from diesel vehicles are higher, especially most OVOC species.

The sentences in the Section 3.3 (line 405-408) are modified to:

Generally, similar variability is obtained except the determined slope of the data points, with higher slopes determined from the scatterplot based on fuel-based emission factor (0.19 versus 0.15). The emission ratios to CO between gasoline and diesel vehicles (Fig. S11b) show similar results.

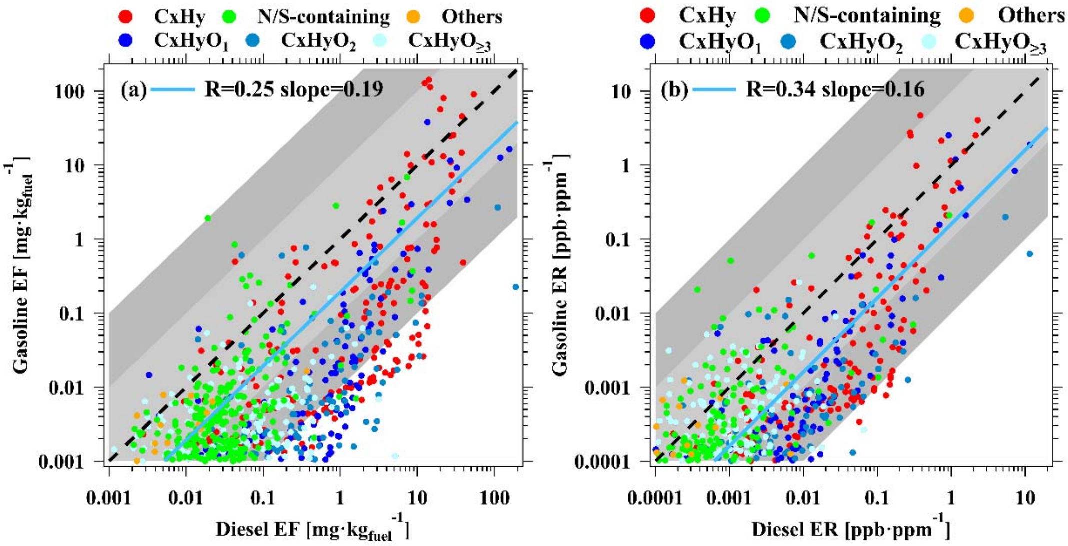

Figure S11. Scatterplot of (a) the determined average fuel-based emission factors (mg·kg$_{fuel}^{-1}$) and (b) the emission ratios to CO (ppb·ppm^{-1}) of VOCs between gasoline and diesel vehicles. Each data point indicates a VOC species measured by PTR-ToF-MS. The blue line is the fitted result for all data points. The black line represents 1:1 ratio, and the shaded areas represent ratios of a factor of 10 and 100.

10. Figure S6. The intercomparisons are nice for the fast time-resolution systems, but there are significant differences between the GC and PTR for toluene - is this due to differences in sampling techniques (e.g., grab sampling artifacts vs real-time sampling), or something due to fragmentation in the PTR to produce a signal at m/z 93? I believe the other reviewer also commented on this, and I agree that some explanation is warranted here.

Reply: We thank the reviewer for the comment. In replying to the comments from the two reviewers, we found an issue in data analysis for preparing the original manuscript. The alignment of data points between offline canister-GC-MS/FID and PTR-ToF-MS for several gasoline vehicles and diesel vehicles was not correct. We have modified the corresponding data points, and added comparison of C_8 aromatics between two measurements (Fig. S6), obtaining generally consistent results, considering large variations of VOC emissions for driving conditions and the difficulty to control the fill time for canisters. We also revised related figures (Fig. 12 and Fig. S12) and description in the manuscript on the fractions of OVOCs in total VOC emissions in various types of vehicles. These modifications do not change any conclusion in the manuscript.

The sentence in the Section 2.3 (line 194-197) is modified to:

We compared emission factors from PTR-ToF-MS and the offline canister-GC-MS/FID (Fig. S6c-d), obtaining generally consistent results, considering the large variation of VOC emissions for driving conditions and the difficulty to control the fill time for canisters.

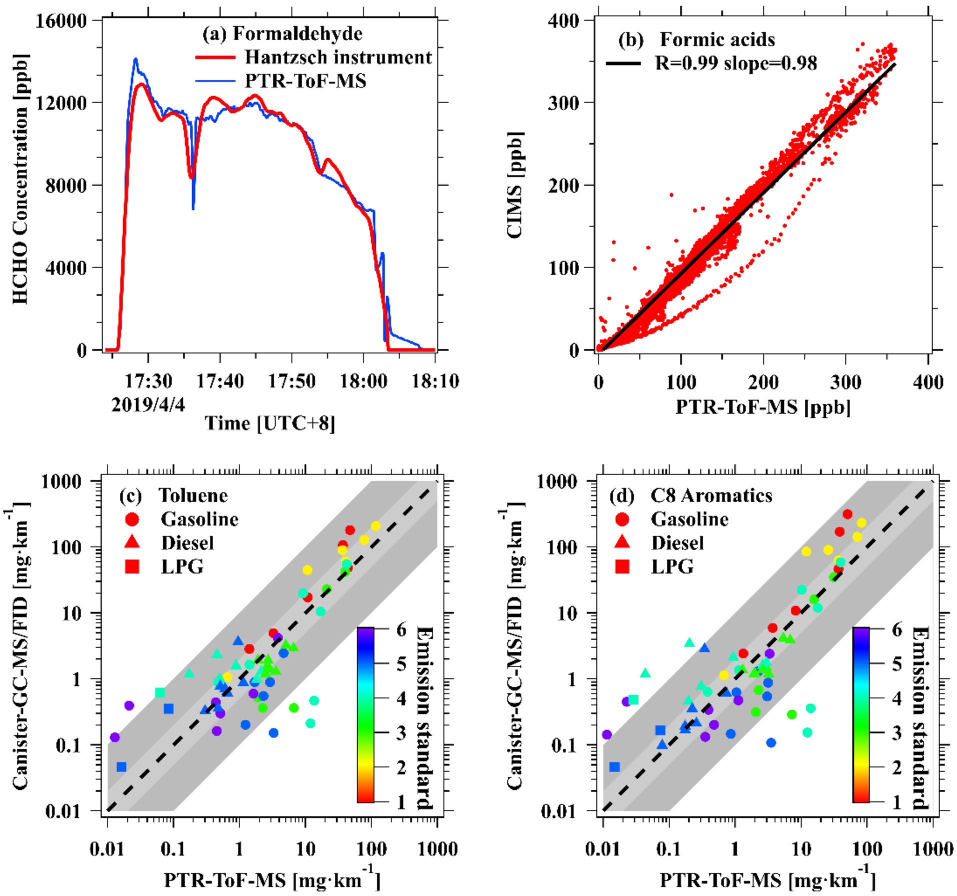

Figure S6. (a) Time series of formaldehyde measured by PTR-ToF-MS and the Hantzsch instrument. (b) Scatterplot of the concentration of formic acid between PTR-ToF-MS and the CIMS. Scatterplot of the emission factor of (c) toluene and (d)C_8 aromatics calculated by the data detected by PTR-ToF-MS and Canister-GC-MS/FID. The black dashed lines represent 1:1 ratio, and the shaded areas represent ratios of a factor of 2 and 10 in (c) and (d).

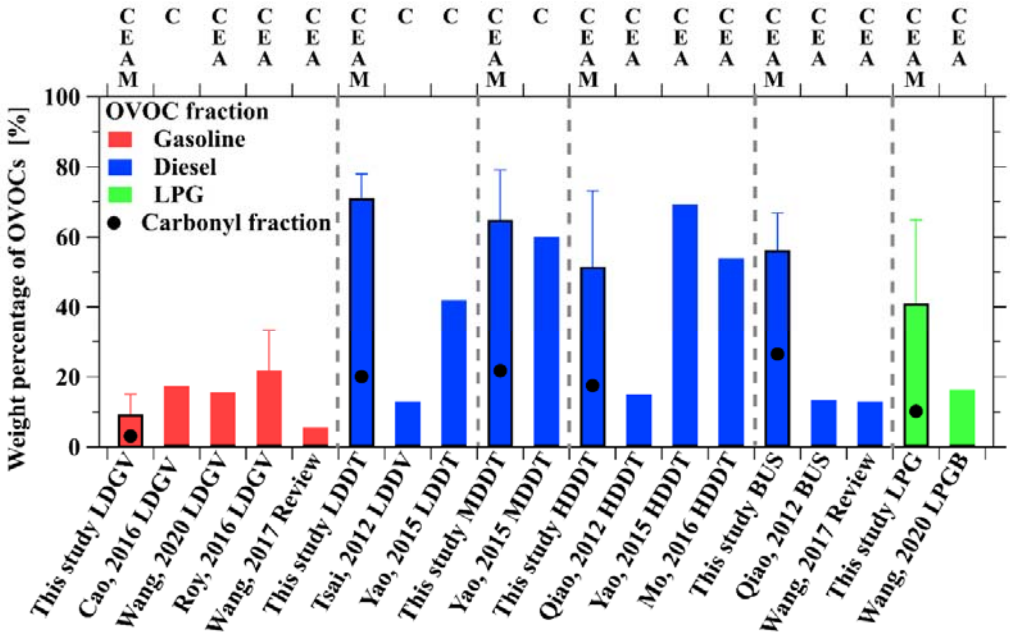

Figure 12. Comparison of OVOCs fractions determined in this study and those in previous studies. Error bars represent the standard deviations of the weight percentage of OVOCs. The C, E, A, M above the top axis represent the four groups of OVOCs measured in this study or previous studies, including Carbonyl: C, Ester/Ether: E, Alcohol: A, Multiple-functional: M.

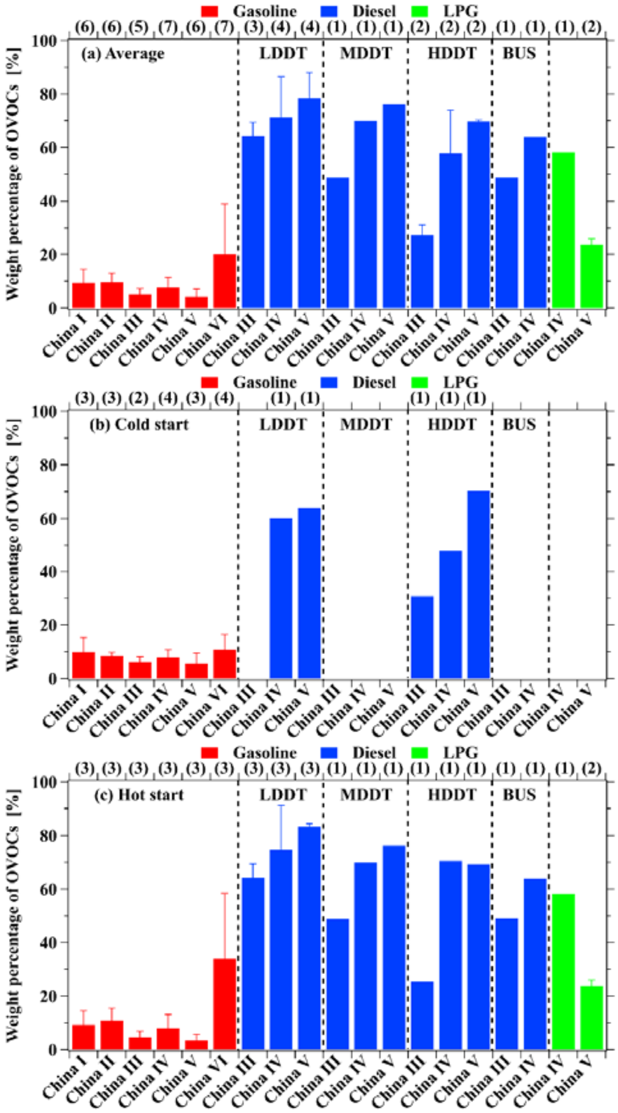

Figure S12. (a) Average OVOC fractions for vehicles with different emission standards, and some difference between (b) cold start and (c) hot start. Error bars represent the standard deviations of the fraction of OVOCs.

Reference:

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